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Investigation of the nonresonant dot-cavity coupling in two-dimensional photonic crystal nanocavities

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We study the optical emission from single semiconductor quantum dots coupled to the optical modes of photonic crystal nanocavities. For dots that are both spectrally and spatially coupled, autocorrelation measurements reveal efficient single photon generation, with a drastically reduced lifetime due to the Purcell effect. However, the multiphoton emission probability is enhanced compared to the same quantum dot (QD) transition when it is detuned from the cavity mode by controlled N_2 deposition. This indicates the presence of an emission background that is shown to be related to the dot by using photon cross-correlation spectroscopy. Photon temporal correlations persist even for large spectral detunings beyond $\Delta\lambda \sim -10$ nm, excluding the intrinsic QD continuum and phonon mediated processes as being responsible for the cavity mode emission background. We propose a mechanism based on photon induced shake up processes in the charged quantum dots, enhanced by the optical cavity.

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Single photon sources based on semiconductor quantum dots[1,2] (QDs) are needed for many applications ranging from quantum key distribution[3] to quantum information processing by using linear optical components.[4] In the latter case, the single photons should be emitted not only very efficiently[5] but also over time scales that are short compared to exciton dephasing processes.[6] This requirement can be fulfilled by incorporating QDs into solid state optical nanocavities and by using the Purcell effect to enhance efficiency and improve photon quantum indistinguishability.[2,7–10] Although the coupling of single quantum dots to the optical modes of such nanocavities has already been achieved by a number of groups worldwide,[9–12] the majority of single dot experiments report a pronounced background emission in the vicinity of the cavity mode, even when a single QD transition is *not* resonantly coupled. Recently, the emission from this background has been shown to be correlated with the emission from the detuned QD, indicating that a mechanism exists by which a spectrally detuned dot can emit into the cavity mode. Several mechanisms have been proposed to account for this emission background, including wetting layer QD continuum transitions,[10] and the presence of correlations has been suggested to be due to phonon assisted processes.[9] However, no systematic investigations have appeared until now and conclusive statements about the origins of the background emission remain elusive.

In this Rapid Communication, we present comparative investigations of single photon generation from self-assembled $In_{0.5}Ga_{0.5}As$ QDs in a photonic crystal (PC) defect nanocavity as a function of spectral detuning between the dot and the cavity mode. By performing auto- and cross-correlation spectroscopy, we examine the influence of the modified photonic environment on the spontaneous emission dynamics of individual dots and prove single photon generation. We find that QD transitions that are spectrally in resonance with the nanocavity mode exhibit a significantly enhanced multiphoton emission probability due to background emission into the cavity mode.[9,10] By controlled adsorption of molecular nitrogen into the PC nanostructure,[13] we detune the cavity mode, enabling us to systematically investigate the influence of the local photonic environment on both the optical properties of the same QD transition and the nature of this background emission. Photon cross-correlation measurements show that all emission lines investigated stem from the same dot. Most remarkably, pronounced cross correlations are observed between the dot transitions and the cavity mode even for very large energy detunings (>12 meV). This surprising observation indicates the presence of an intrinsic coupling mechanism between the spectrally detuned QD and the cavity mode.

The samples studied are grown by molecular beam epitaxy and consist of the following layers grown on a semi-insulating GaAs wafer: an undoped GaAs buffer followed by a 500 nm thick $Al_{0.8}Ga_{0.2}As$ sacrificial layer. This was followed by an 180 nm thick GaAs waveguide at the midpoint of which a single layer of self-assembled $In_{0.5}Ga_{0.5}As$ QDs was incorporated. A two-dimensional (2D)-PC was formed by patterning a triangular array of cylindrical air holes by using electron-beam lithography and reactive ion etching. The lattice constant of the PC was $a=280$ nm and the air hole radius $r=0.33a$. Nanocavities were established by introducing three missing holes to form an *L*3 cavity.[14] Finally, freestanding GaAs membranes were formed by an HF wet etching step. A scanning electron microscopy image of the investigated nanocavity is shown in the inset of Fig. 1(a) and an overview of the different fabrication steps can be found in Ref. 15.

The sample was mounted in a liquid He-flow cryostat and cooled down to $T=15$ K. For excitation, we used either a pulsed Ti:sapphire laser ($f_{laser}=80$ MHz, 2 ps duration pulses) or a continuous wave (cw) laser tuned into the wetting layer (WL) continuum at $\lambda_{exc}=837$ nm. The QD microphotoluminescence (μ-PL) was collected via a 100× microscope objective (numerical aperture=0.8) providing a spatial resolution of ~700 nm and the signal was spectrally analyzed by a 0.55 m imaging monochromator and detected with a Si-based, liquid nitrogen cooled charge coupled device detector. For time-resolved measurements, we used a fast silicon avalanche photodiode that provided a temporal resolution of ~100 ps after deconvolution. A pair of similar

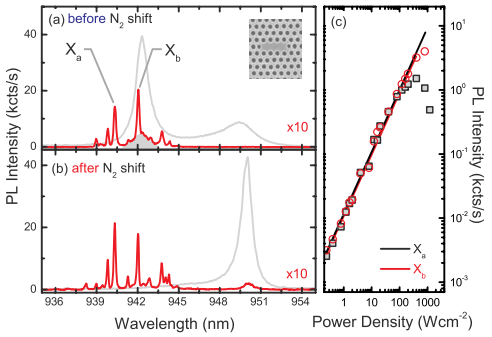

FIG. 1. (Color online) (a) μ-PL spectra recorded from a single QD coupled to a cavity mode for weak (red/gray) and strong (gray) cw pumping. The gray shaded region indicates the mode background emission. (Inset) Scanning electron micrograph of the $L3$ cavity investigated. (b) μ-PL spectra of the same QD and the spectrally shifted cavity mode after N_2 deposition. (c) PL intensity as a function of the excitation power for the X_a and X_b lines.

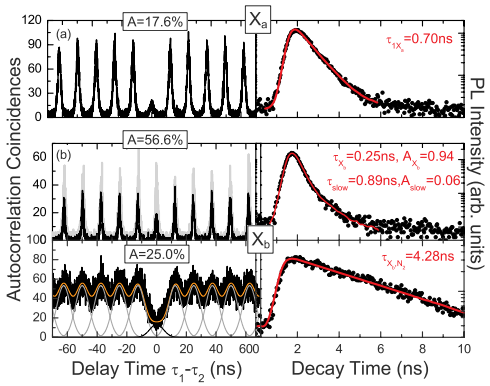

FIG. 2. (Color online) (Left panels) Autocorrelation measurements and corresponding time-resolved data (right panels) for (a) X_a and (b) X_b. The two panels in (b) compare the data for X_b coupled to the cavity mode before N_2 deposition with the case when X_b is spectrally detuned from the cavity mode after N_2 deposition. An autocorrelation of the mode background is shown in gray.

detectors in Hanbury Brown and Twiss configuration were used for both photon auto- and cross-correlation measurements.

In Fig. 1(a), we present μ-PL measurements of a single QD that is spectrally and spatially coupled to a PC nanocavity. For weak cw excitation (0.4 W/cm^2), we observe several emission lines, all stemming from different transitions of the same QD [Fig. 1(a), red/gray trace]. The two most prominent lines, labeled X_a (λ_{X_a}=940.33 nm) and X_b (λ_{X_b}=942.04 nm) in the figure, are *out of resonance* and *in resonance* with the cavity mode ($\lambda_{cav,0}$=942.32 nm). The cavity mode can be clearly observed at higher excitation powers (400 W/cm^2) and has a quality factor of 850 [Fig. 1(a), gray trace]. Both X_a and X_b show a linear power dependence [Fig. 1(c)] demonstrating that they arise from single exciton transitions. Cross-correlation measurements between X_a and X_b reveal that both emission lines arise from the same QD (presented below) probably due to different charge states. When compared to X_a, transition X_b saturates at higher excitation power, as can clearly be seen by examining Fig. 1(c). This is a clear sign of the Purcell enhanced emission due to the spectral proximity of the cavity mode,[16] a hypothesis tested below by using time-resolved spectroscopy. To investigate the influence of the cavity on the emission, we spectrally shifted λ_{cav} by using a controlled N_2 deposition technique.[13] A typical result is shown in Fig. 1(b), where λ_{cav} has been detuned by $\Delta\lambda=+7.7$ nm to λ_{cav,N_2} =950.2 nm. This procedure allows us to investigate the *same* QD state X_b when it is spectrally coupled to the cavity mode (*before* N_2 deposition) and when strongly detuned (*after* N_2 deposition). By comparing the (low power) μ-PL spectra in Figs. 1(a) and 1(b), we note that no emission is observed close to λ_{cav,N_2}=950.2 nm before the N_2 shift was executed.[17] This indicates the presence of a background emission associated with the cavity mode as was recently reported in Refs. 9 and 10.

To confirm that we probe transitions of a single QD, we performed photon autocorrelation and time-resolved spectroscopy measurements on X_a and X_b when subjected to pulsed optical excitation. These measurements were obtained with a $\Delta\lambda=0.3$ nm wide detection window centered on the transition of interest and the results are presented in Fig. 2(a) for transition X_a and Fig. 2(b) for X_b. The upper and lower panels of Fig. 2(b) separately compare the measurements recorded from X_b before and after the N_2 shift was executed. For X_a, which is initially detuned by $\Delta\lambda=+2$ nm from the cavity, we observe clear triggered single photon generation with a multiphoton emission probability of $P_{X_a}=17.6\%$. The spontaneous emission lifetime is $\tau_{X_a}=0.7\pm0.1$ ns [Fig. 2(a)], slightly larger than the typical lifetime of dots in unpatterned bulk GaAs for this sample ($\tau_{bulk}=0.6$ ns) but much shorter than the typical lifetime of dots emitting deeply within the photonic band gap [4–12 ns (Ref. 5)]. The multiphoton emission probability of X_b [Fig. 2(b), upper panel] is clearly enhanced ($P_{X_b}=56.6\%$) when compared to X_a, indicating that background emission from the cavity mode also contributes to the measured count rate within our detection bandwidth. The mode background, on the other hand, shows no antibunching at all but rather Poissonian distributed light, as shown in gray in Fig. 2(b) (upper panel). This conclusion is not only supported by the observation that X_b is superimposed on a broadband emission [gray shaded region in Fig. 1(a)], which is absent after N_2 deposition [Fig. 1(b)], but also by autocorrelation measurements of the same transition after N_2 deposition [Fig. 2(b), lower panel]. Due to the considerably longer lifetime $\tau_{X_b}^{N_2}=4.28\pm0.13$ ns when the mode is detuned, the peaks in the autocorrelation spectrum broaden leading to a quasi-cw-like result since $\tau_{X_b}^{N_2}\approx 1/f_{laser}$. As for the in-resonance case, discussed above, we observe pronounced photon antibunching although with a smaller multiphoton emission probability of $P_{X_b}^{N_2}=25.0\%$ compared to the value of $P_{X_b}=56.6\%$ before the N_2 deposition. This clearly indicates that shifting the mode away from the dot enhances the purity of single photon emission.

Further insights into the dot-cavity system investigated and the nature of the background emission are obtained by examining the time-resolved data presented in Fig. 2(b). Detecting on X_b, we observe a biexponential decay transient before N_2 deposition but a clear monoexponential decay af-

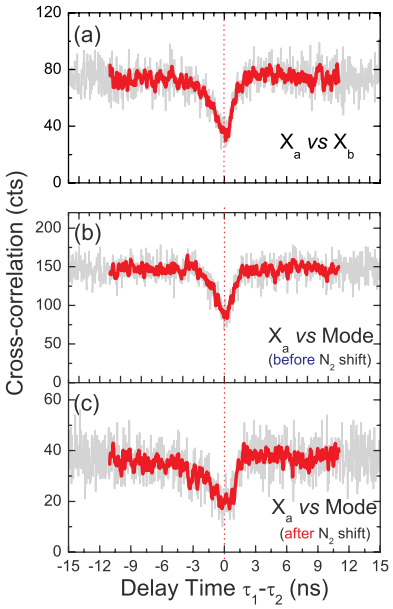

FIG. 3. (Color online) Cross-correlation measurements between (a) X_a and X_b, (b) between X_a and cavity mode before N_2 deposition, and (c) between X_a and cavity mode after N_2 deposition. We show the original data in gray and the corresponding smoothed data in red/gray.

terwards. We attribute the faster component of the biexponential decay $\tau_{X_b}=0.25\pm0.1$ ns to the Purcell enhanced emission of the coupled X_b transition and the slower lifetime ($\tau_{slow}=0.89\pm0.1$ ns) to the background emission into the cavity mode. Further support for this conclusion is provided by analyzing the lifetime-amplitude products of the fast and slow components of the decay, namely, $\frac{A_{slow}\tau_{slow}}{A_{X_b}\tau_{X_b}}\sim 23\%$. This value compares very well with the measured ratio of the intensities of X_b and the mode background emission $\frac{PL_{back}}{PL_{X_B}}\sim 22\%$ observed in μ-PL (Fig. 1), confirming the identification of the fast and slow time constants. Furthermore, by comparing τ_{X_b} measured before and after the N_2 shift, we estimate the degree of spatial coupling of the QD to the cavity mode to be $\left|\frac{E(\vec{r})}{E_{max}}\right|\geqslant 40\%$,[18] demonstrating that we deal with a dot-cavity system that is spectrally and spatially well coupled. Since we spectrally select only one of the QD emission lines, namely, X_b, this provides further support that the cavity mode background is responsible for the enhanced multiphoton emission probability observed in Fig. 2 before the N_2 shift.

To prove that the emission from X_a and X_b stem from the same quantum emitter and to further probe the nature of the cavity mode background, we performed cross-correlation measurements. In Fig. 3(a), we show the cw cross-correlation measurement between X_a and X_b before the N_2 shift was performed.[19] A pronounced dip is observed at $\tau_1-\tau_2=0$, clearly showing that both transitions stem from the same QD, the different lifetimes of the two quantum states giving rise to the observed asymmetric character of the dip.[20] We also performed cross-correlation measurements between X_a and the mode background emission, beside X_b at 943.0 nm [Fig. 3(b)]. The clear dip in the cross-correlation histogram [Fig. 3(b)] unambiguously shows that the background emission from the mode arises from the same QD. Most remarkably, we measure a similar cross correlation between X_a and the cavity mode after N_2 deposition when the mode has been shifted by +7.7 nm to longer wavelengths. Anticorrelations were found to persist as the detuning was systematically increased from <2 up to >10 nm.[21] This implies the existence of an intrinsic dot-cavity coupling mechanism that is active for large dot-cavity detunings, which clearly cannot be explained by the simplified atomlike picture that has been so successfully applied to quantum dots up to now.

While the precise mechanism responsible for nonresonant dot-cavity coupling is not yet fully clear, the observation already indicates the presence of energy exchange with a *continuum reservoir*. Most importantly, we can already exclude a number of possibilities that have been discussed in the literature: Emission from 2D-2D WL states, crossed (continuum) 0D-2D transitions,[22,23] and excited state QD transitions can all be excluded since they result in emission at higher energy than the fundamental exciton transition in the dot. Only mechanisms that lead to a *reduction* in energy, and emission of a photon with *lower* energy than the fundamental transition, could contribute to the emission of the cavity mode in our scenario. Recently, Press *et al.*[9] suggested that the nonresonant coupling might be mediated by the emission of acoustic phonons. Such processes are extremely unlikely for large detunings, since the exciton-acoustic phonon coupling strength is strongest for phonon wave vectors $q_{ph}^{max}\sim\pi/d$, where d is the characteristic confinement length scale in the dot. For $E_{ph}\sim 10$ meV, the phonon wave vector is $q_{ph}=E_{ph}/v_s\hbar\sim 10q_{ph}^{\max}$ and we estimate that the acoustic phonon coupling strength is more than 6 orders of magnitude weaker as compared to the case for $q_{ph}\sim q_{ph}^{\max}$.[24] We have observed cross correlations for energy detunings in excess of 19 meV (cf. Ref. 21), excluding acoustic phonon mediated processes.

We discuss a possible mechanism that is consistent with all our experimental observations and those of other groups.[9,10] QDs in the PC exhibit more complicated μ-PL spectra compared to QDs in the bulk (not shown here), which indicate the existence of charged dots, possibly due to charge trapping at the etched GaAs surface in the PC.[25] Thus, it is feasible that we could encounter a scenario where charged exciton decay takes place into a *continuum* of final states. This idea is illustrated in Fig. 4 for the case of a dot charged with n holes. In the initial state of the transition, labeled $|i\rangle$ in Fig. 4, all particles are accommodated by the orbital states of the dot due to the balance between attractive and repulsive Coulomb interactions in the X^{n^+} charged exciton initial state. However, in the n-hole final state of the transition, configurations exist where holes are distributed between the dot and the WL continuum, labeled by $|f^{con}\rangle$ in Fig. 4. The absolute energy of this final state continuum would be determined by the interplay between the repulsive Coulomb interaction in the n-hole final state and the kinetic energy cost of ejecting particles into the continuum.[26] Exciton decay can then take place into discrete and continuous final states, with the branching ratio depending on the relative oscillator strength for each state. We suggest that decay

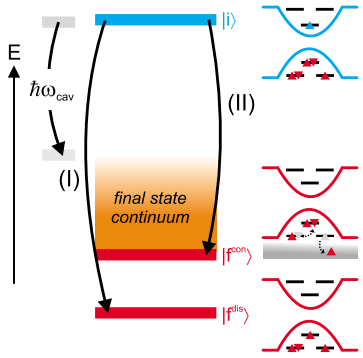

FIG. 4. (Color online) Energy diagram with initial state $|i\rangle$ and two final state configurations $|f^{con}\rangle$ and $|f^{dis}\rangle$ and the corresponding QD occupations for a heavily positively charged dot.

into such a delocalized final state continuum may be enhanced close to the cavity energy giving rise to the observed behavior. This process would explain the observed background emission in the cavity mode and also explain both the different decay lifetimes measured for the dot and cavity and the observation of correlations between the photons emitted (X^{n+}) state. We note that such a nonresonant coupling must occur *during* photon emission, the mechanism being similar to the photoinduced hybridization of quantum levels recently observed for highly charged QDs.[27]

In summary, we presented the investigations of single photon generation and dot-cavity coupling effects for self-assembled QDs in PC defect nanocavities. We have shown that, for dots that are spectrally and spatially coupled to the cavity, single photon generation is observed with elevated multiphoton emission probability. This was shown to be caused by background emission in the cavity mode. By shifting the cavity mode away from the dot by adsorption of molecular N_2 into the PC cavity, we studied photon cross correlations between the dot transitions and the detuned cavity mode. Pronounced cross correlations were observed, which persist even for very large energy detunings (>12 meV). We note that this demonstration of nonresonant interaction is in the weak coupling regime and supports the findings of Refs. 9 and 10 that were both in the strong coupling regime. We suggest that the coupling between QD and mode is mediated by photon mediated shake-up-like processes in which the final state of the transition is an excited continuum.

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